

03500.017559.

PATENT APPLICATION

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of:	)	
	:	Examiner: Rita J. Desai
MAKI OKAJIMA ET AL.	)	
	:	Group Art Unit: 1625
Application No.: 10/527,192	)	
	:	Confirmation No.: 1113
Filed: March 10, 2005	)	
	:	
For: PHENANTHROLINE	)	
COMPOUND AND ORGANIC	:	
LIGHT EMITTING DEVICE	)	
USING SAME	:	
	)	

Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

DECLARATION UNDER TITLE 37 C.F.R. § 1.132

Sir:

I, KOICHI SUZUKI, declare that:

1. I reside at 3-3-201, Ohdananishi, Tsuzuki-ku, Yokohama-shi,  
Kanagawa, JP.
2. I have been a Research Scientist at Canon Kabushiki Kaisha since  
1986. Prior to my employment at Canon I held the positions of master's course  
Tokyo Agriculture and  
at Technology University during the period. I have worked in the areas of research and  
development of organic light-emitting diodes and in developing organic photoconductors.

3. I have received a graduate degree from Tokyo University and a doctoral degree in engineering from Saga University.

4. I have received the following patents in my name: U.S. Patent Nos. 6,458,476; 6,461,748; 6,461,749; 6,652,997; 6,916,555; 6,994,922; 7,241,513 and 7,338,721.

5. I have co-authored the following articles: (1) "Synthesis of Fluorescent 1,2-Oxazole Compounds", Koichi Suzuki and Kazunori Ueno, *Journal of Photopolymer Science and Technology*, 14, 311 (2001). (2) "Simple synthesis of Ring-fused pyridazin-3-ones", Koichi Suzuki, Akihiro Senoh and Kazunori Ueno, *Heterocycles*, 57, 723 (2002). (3) "New Host Materials for Blue Emitters", Koichi Suzuki, Akihiro Senoh, Hiroshi Tanabe and Kazunori Ueno, *Synthetic Metals*, 143, 89 (2004).

6. I am an inventor of the subject patent application and am familiar with the prosecution history of the subject patent application.

7. The Examiner relies on the disclosure of 2,9-difluorenyl-bathophenanthroline in Example 4 of U.S. Patent No. 6,972,334 B1 issued to Shibamura, et al and its equivalent, EP 1,097,980, in paragraph [0094] thereof, also known as RN# 338734-80-8 (1,10-Phenanthroline, 2,9-di-9H-fluoren-9-yl-4,7-diphenyl).

8. I tried to reproduce the synthesis of 2,9-difluorenyl-bathophenanthroline (Compound X) in Example 4 of U.S. Patent No. 6,972,334 B1 and Example 4 of corresponding EP 1,097,980 in paragraphs [0094-0098]. Since the origin of the lithium diisopropylamine (LDA) used in Example 4 was not identified, I prepared LDA by a conventional reaction of diisopropylamine and n-butyllithium. I also made several minor changes in amounts of solvent and starting

material and in reaction time to expedite the synthesis. In particular, I employed half the amounts of fluorene and bathophenan-throline. Any such changes were not sufficient to adversely effect the synthesis. In the following synthesis procedure for compound X I have noted in brackets where a change was made from the reported synthesis for the Examiner's convenience:

1.4 ml (10.0 mmol) of diisopropylamine and 30 ml of THF were placed in a 200 ml three-necked flask, and 5.4 ml (8.34 mmol) of n-butyllithium was added dropwise thereto at 0°C with stirring in a nitrogen atmosphere. The mixture was heated to room temperature and stirred for 3 hours to give lithium diisopropylamine (LDA).

Then, 30 ml of a THF solution of 2.0 g (12.0 mmol) [4.16 g, 25.0 mmol] of fluorene was added into the flask at 0°C with stirring and the mixture was heated to room temperature and stirred for 12 [16] hours. Thereafter, the reaction solution (THF and diisopropylamine) was evaporated under reduced pressure. To 20 ml of an anhydrous diethyl ether solution of the resulting yellow solid, 60 ml of a toluene solution of 1.0 g (3.0 mmol) [2.63 g, 6.11 mmol] of bathophenanthroline was gradually added dropwise at 0°C [room temperature] with stirring. After the dropwise addition, the mixture was heated to reflux for 3 [2] hours and the reaction solution was stirred at room temperature for 8 [16] hours.

Ice-cold water was gradually added to the resulting reaction solution to separate an organic layer. The aqueous layer was extracted three times with chloroform, and the resulting organic layer was mixed with the previously separated organic layer. 60 g of manganese dioxide was added to the thus mixed organic layer and the mixture was

stirred for 30 minutes, after which 100 g of sodium sulfate was further added, followed by stirring for 30 minutes.

The resulting mixed solution was filtered and concentrated. When the residue was spotted on a silica gel column chromatography (TLC) plate and the plate was immersed in toluene developing solvent, an unknown spot migrated to a position which was intermediate between the spots of bathophenanthroline and fluorene as the starting materials on the thin layer chromatogram, as shown on the attached Exhibit entitled "(1) Retry of Ex. 4 in EP 1097980 (K. Suzuki No. 2)". It is presumed that the unknown spot is attributable to Compound X. Therefore, the residue was purified through silica gel column chromatography (chloroform/heptane mixed developing solvent), followed by recrystallization (solvent for recrystallization: IPE) to obtain the intended compound (0.35 g) which showed the above-mentioned spot migration.

When the product was measured through  $^1\text{H-NMR}$ , as illustrated on the attached Exhibit 2 entitled as "(2) Identification of unknown sample from Ex. 4 (K. Suzuki No. 3)", protons were not detected at 4.51 where benzyl protons of Compound X should be measured as (m, 2H, Ar-CH<sub>2</sub>-Ar). Further, when the product was measured through Maldi TOF MS, as illustrated in Exhibit 2, 660 which should be observed as MS of Compound X was not detected.

9. I have requested other experts who are colleagues of mine to attempt to synthesize Compound X by various synthesis methods including (a) the Suzuki coupling method using a palladium catalyst (Chem. Rev. 1995, 95, 2457-2483 and (b) a phenanthroline compound intermediate as in J. Org. Chem., 16, 941-945 (1951), but they were unsuccessful.

10. It is my expert opinion that there was no available synthesis for preparing Compound X of Example 4 of EP 1,097,980 as of its publication date of May 9, 2001.

11. I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Dated: September 3, 2008

Koichi Suzuki

KOICHI SUZUKI